AN ACOUSTIC TECHNIQUE FOR THE NONINVASIVE IN-SITU MEASUREMENT OF CRYSTAL SIZE AND SOLUTION CONCENTRATION

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We demonstrated the use of acoustic measurements for tracking potassium dihydrogen phosphate (KDP) crystal growth. Both KDP solution concentration and KDP crystal size can be found by using information derived from acoustic wave propagation in the solution. Acoustic measurements show good correlation to conductivity measurements for KDP solution concentration.

INTRODUCTION

The National Ignition Facility (NIF) at Lawrence Livermore National Laboratory (LLNL) is currently developing rapid growth techniques for potassium dihydrogen phosphate (KDP) single crystals. NIF requires over six hundred KDP optical components, sized about 0.4 meters square, for optical switching and frequency conversion of the beam line. Rapid boule growth rates of approximately 10-15 mm/day are necessary to produce the large number of KDP optical components at a reasonable cost. While conventional growth methods would take more than 1 year to grow a KDP crystal of the size required, NIF scientists are developing a technique to grow the crystal in approximately 8-10 weeks.

Crystals are grown from a point seed in a highly supersaturated solution of KDP. The supersaturation level of the solution is the primary operational parameter used to determine the growth rate of the crystal.[1] Thermal control of KDP solution is used to keep supersaturation at desired levels. Fast growth rates of such large crystals requires stringent controls on the KDP solution concentration.

Several techniques are commonly used to determine solution concentration in crystal growth. The current method uses visual measurement of the KDP crystal size. Given the known solid density of the KDP crystal, the mass of salt removed from solution is estimated. Subtraction of this estimated value from the initial starting salt gives the solution concentration as a function of time. This method is rather inaccurate because

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visual crystal sizing is accurate to only 1-2 mm. Other methods using density (from buoyancy), refractive index and electrical conductivity are also commonly used commercially by crystal growers to determine solution concentration. Generally, solution concentration is determined from empirical relationships of the measured properties. These commercial techniques all require access to the solution. Acoustic measurements offer the possibility of a noninvasive technique for monitoring crystal growth. High frequency acoustic measurements have the advantage of noninvasively obtaining information about the solution concentration and crystal size during the growth process.

This work assessed acoustic velocity as a diagnostic for KDP crystal growth. Electrical conductivity measurements for solution concentration are used as a comparison. Acoustic velocity measurements are also tested as a means of sizing the crystal during growth.

PROGRESS

We designed a probe to empirically determine the relationship between acoustic velocity and temperature and concentration of KDP solution. Used for the purpose of determining the empirical relationship, this probe is designed to operate invasively in the solution. After determining the empirical relationships, acoustic measurements can be made noninvasively from outside of the solution.

Fig. 1 shows a photograph of the probe and the measured acoustic waveform. A 15 MHz ultrasonic transducer mounted on the top of the probe sends a wideband pulse (bandwidth = 55 %) propagating into the probe. Fabricated from fused silica, the probe has threaded sides to disperse diffracted waves incident on the sides.

Notched into the probe is a 10 mm gage length. The low coefficient of thermal expansion of fused silica ensures that the gage length remains constant through the temperature range of KDP supersaturation. A pulsed acoustic wave travels roundtrip from the transducer, into the probe, through the solution within the gage length and back to the transducer through the probe. Travel time of the pulse through the KDP solution gives the acoustic velocity. The wideband 15 MHz ultrasonic pulse permits high temporal resolution of the acoustic velocity in the KDP solution. The probe can discern changes as small as 0.25% in acoustic velocity.

In fluids, acoustic velocity is directly related to density. For example, empirical relationships in sea water show that acoustic velocity increases linearly with salinity and as a second order polynomial with temperature. [2] KDP solution should follow acoustical relationships similar to those found in sea water.

Fig. 2 shows acoustic velocity and conductivity data for different concentrations of KDP solution. Curve fitting indicates that acoustic velocity is related to temperature through a second order polynomial. At a given temperature, velocity increases with solution concentration. The data show that acoustic velocity peaks at approximately 60°C - 65°C for all concentrations. From linear regression at T = 60°C

$$v(C_0) = 0.6414C_0 + 1.534 \qquad (1)$$

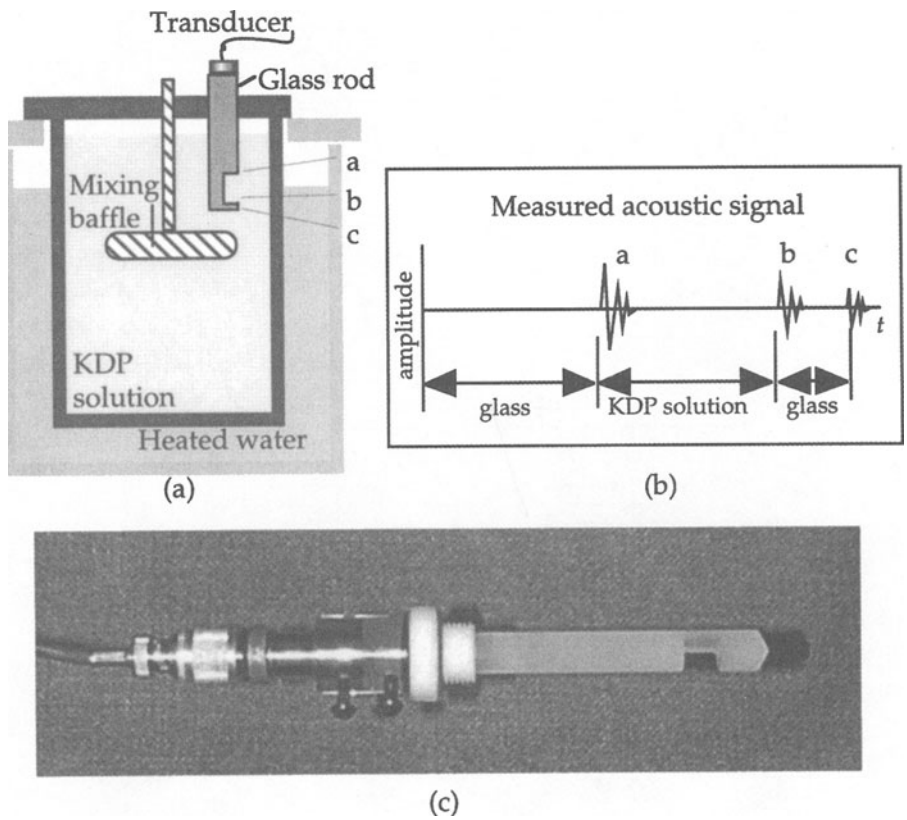
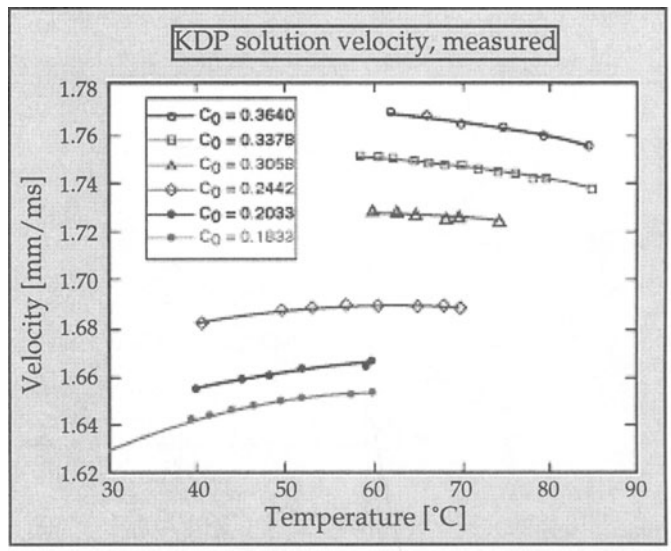
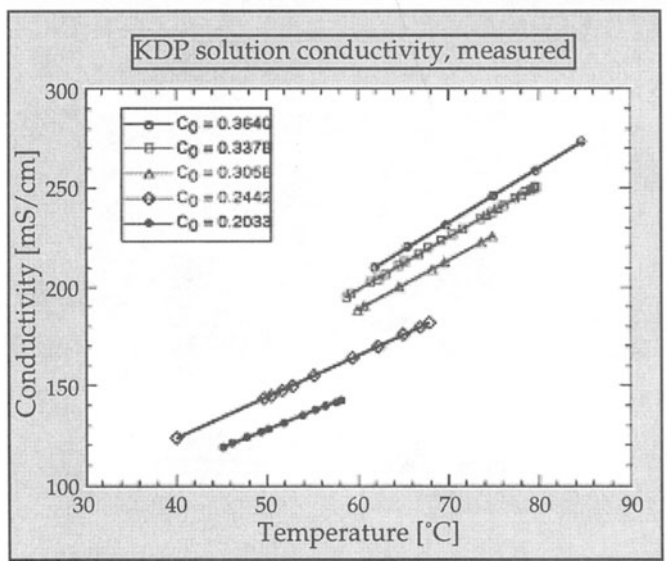


Fig. 1. Attached to a 15 MHz transducer, the fused silica probe with 10 mm gage length is placed in the KDP solution to measure acoustic velocity. (a) Measurement configuration, (b) Measured acoustic signal,(c) Photograph of the probe. Three reflections from the probe are measured in the signal. Velocity is found from the travel time in the KDP solution.



(a)



(b)

Fig. 2. Acoustic velocity (a) and conductivity (b) are measured at different temperatures and concentrations. For a given concentration, velocity follows a second order polynomial fit while conductivity increases linearly with temperature.

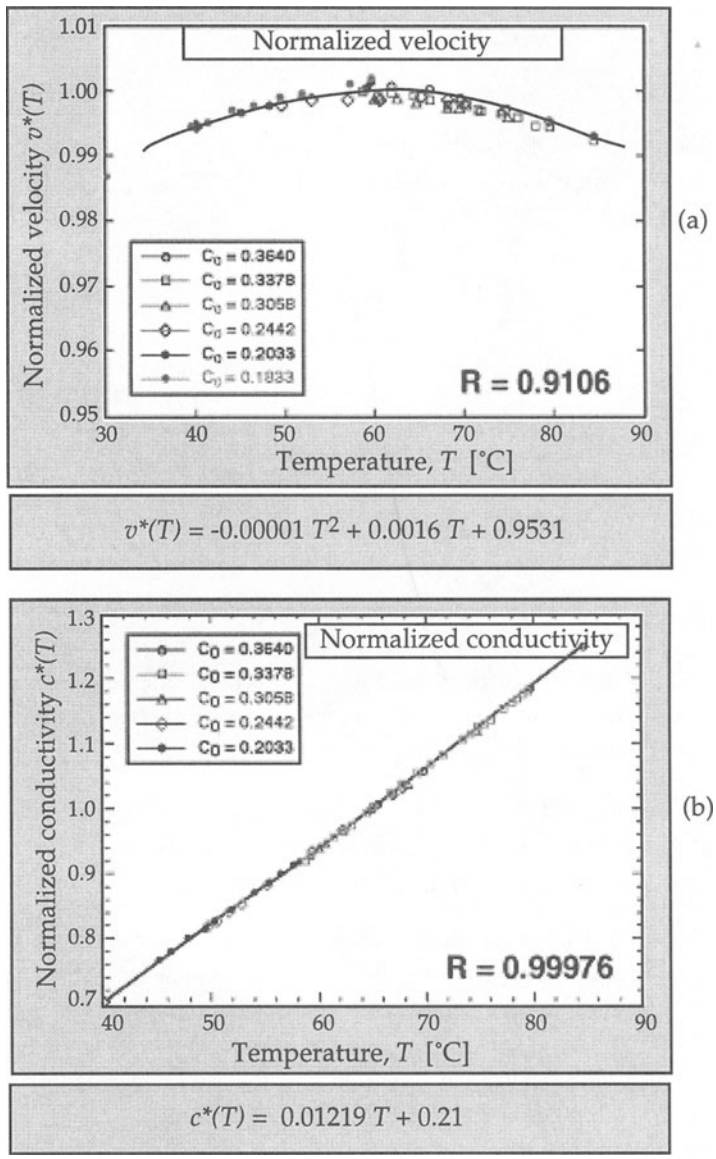


Fig. 3. Empirical equations are derived for normalized velocity (a) and normalized conductivity (b). Velocity and concentration are normalized for concentration according to eqn. (1) and eqn. (2). Velocity follows a second order polynomial fit with temperature with the maximum velocity occuring at 60 °C. Conductivity fits its empirical equation (R = 0.99976) better than velocity (R = 0.9106).

where C_0 = solution concentration and $v(C_0)$ =acoustic velocity.

For a fixed concentration, conductivity increases linearly with temperature, as expected. At T = 65°C,

$$c(C_0) = 379.7C_0 + 82.9 \tag{2}$$

where $c(C_0)$ =conductivity.

Empirical equations derived from curve fitting for normalized velocity and concentration are shown in Fig. 3. In Fig. 3a, measured velocity, $v_{measured}$, is normalized to $v(C_0)$ by the equation

$$v^*(T) = \frac{v_{measured}}{v(C_0)} \tag{3}$$

where $v^*(T)$ is normalized velocity. In Fig. 3b, conductivity is normalized to $c(C_0)$ by the equation

$$c^*(T) = \frac{c_{measured}}{c(C_0)} \tag{4}$$

where $c^*(T)$ is normalized conductivity.

Conductivity shows better correlation to concentration (R = 0.99976) than velocity (R = 0.9106), though for some applications, the non-intrusive nature of the acoustic technique may be of more importance than the small loss of accuracy of the measurement. Also, the conductivity probe is very sensitive to the presence of bubbles on its measurement surface, which strongly affect the conductivity reading.

The empirical equations shown in Fig. 3 are useful for in-situ continuous monitoring of the solution concentration. Given a temperature, these equations allow us to relate measured values of velocity and conductivity to concentration.

Crystal size in solution is currently measured visually with a telescope and a scale mounted to the side of the tank. The resolution currently obtainable is approximately 1-2 mm. Acoustic measurements of crystal size were performed with the configuration sketched in Fig. 4. The arrival time of the reflected wave from the crystal face is calibrated to the arrival time of the reference wave. This technique is sensitive to approximately 0.1 mm changes in crystal size, resulting in a factor of 10 improvement.

The configuration in Fig. 4 shows a noninvasive method of measuring solution concentration in addition to crystal size. Acoustic velocity is obtained from the reference signal. Given the temperature and acoustic velocity, the solution concentration can be determined using the empirical relations in Fig. 3.

SUMMARY

Acoustic measurements have proven to be a viable, continuous, in process diagnostic technique for crystal growth. The technique has no adverse effect on the

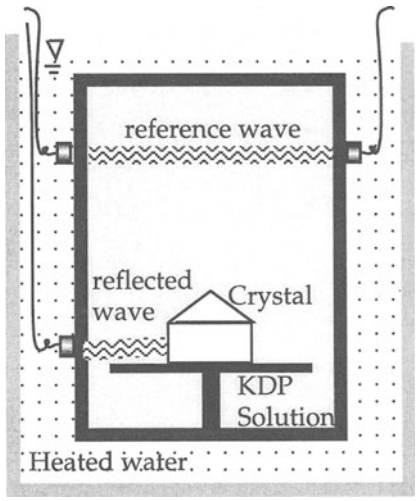


Fig. 4. Acoustic techniques can be used as a noninvasive diagnostic for crystal growth. Solution concentration is found from a reference wave propagating through the solution. Crystal size is derived from the travel time of the reflected wave from the crystal.

solution. Acoustic velocity measurements for solution concentration correlate well with electrical conductivity measurements. This work developed a supplemental tool for the continuous measurement of both KDP solution concentration and crystal size.

FUTURE WORK

Because of its ultimate simplicity and minimal cost, visual observation is currently the method being used to determine crystal size during growth. Solution concentration, however, is currently being measured only before and after the crystal growth run. It would be very desireable to have continuous measurement capability to obtain this quantity throughout the growth run.

This work demonstrated the feasibility of using acoustic measurements for determination of the solution concentration. Additional work is needed, however, to minimize the cost of a fielded acoustic measurement system and to provide a more convenient operator interface for converting the raw acoustic signals to a direct reading of solution concentration.

ACKNOWLEDGEMENTS

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2. Birks, A. S., Green, R. E., "Nondestructive Testing Handbook, Ultrasonic Testing", Vol. 7, 1991.